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Phase transition of quantum spin liquid in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in weak magnetic field to chiral state with topological order during sample magnetization reversal

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In this work, a detailed study of the nature of phase transitions in the Z_2 quantum spin liquid in La $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ manganites induced at a temperature of 4.2 K by a weak external dc magnetic field in the ZFC and FC regimes of measuring the field dependences of the magnetization in the field range of ± 5 kOe is carried out. It is shown that during the magnetization reversal of the sample in the ZFC-regime near the external magnetic field $H = 0$, a second-order phase transition Z_2 quantum spin liquid occurs in the state of a chiral gap quantum spin liquid with a topological order. The transition is accompanied by an unusual hump-like increase in the number of magnetic excitations in the form of Majorana fermions and local gauge Z_2 magnetic fluxes in the range of fields ± 500 Oe near zero field. A characteristic feature of the magnetization features is their strong dependence on the direction of growth of the external magnetic field, which is accompanied by the breaking of the mirror symmetry of the sample magnetization upon reversal of the sign of the external magnetic field, which is typical for the Z_2 chiral state of a quantum spin liquid. In the FC-measurement regime, characteristic signs of excitation-destruction of 1D-fragments of gapless charge/spin density waves were found near $H = 0$ during sample magnetization reversal due to confinement-deconfinement of spinon pairs in a system of spin chains.

Keywords: chiral quantum spin liquid, Majorana fermions, confinement-deconfinement of spinon pairs, 1D-charge/spin density waves.

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1. Introduction

Chiral spin liquids with a topological order parameter form a broad subclass of spin liquids with unusual physical properties that have been intensively studied in two-dimensional frustrated antiferromagnets with various types of crystal lattice over the past decades. A distinctive feature of the chiral state of a spin system is the violation of time symmetry and mirror symmetry [1–12]. Recently, the phase transition of a quantum spin liquid (QSL) to the chiral state in 2D-frustrated AFMs with different types of crystal lattice, caused by the action of an external magnetic field close to $H = 0$ has been of great interest among theoreticians and experimenters. It was shown that the QSL transition to the chiral state induced by an external magnetic field is accompanied by a phase transition into a phase with a topological order and excitation of fractional fermions (Majorana fermions). Kitaev [13] was the first to construct a quantitative model of the so-called Z_2 quantum spin liquid (a spin liquid with a local Z_2 magnetic flux in a lattice cell) for spins $S = 1/2$ located at the nodes of a quasi-two-dimensional hexagonal lattice. The Kitaev Hamiltonian of QSL describes the states of both gap and zero-gap quantum spin liquids, which correspond to low-energy fractional excitations. According to Kitaev's model, there is a strong anisotropic exchange between spins in the nearest surrounding of nodes in a simple Ising form, but due to the fact that different bonds use different spin components, the ground magnetic state of the system becomes highly frustrated. According to the model, the ground state of such a system of spins in a zero magnetic field is a zero-gap quantum spin liquid, which can pass into a gap topological phase as a result of the action of perturbations that break the time reversal symmetry. One of such perturbations can be an external magnetic field perpendicular to the hexagonal layer. In Kitaev's theory, the QSL phase transition to the gap state is accompanied by the excitation of Majorana fermions (MFs). In elementary particle physics, a Majorana fermion is understood as a particle coinciding with its antiparticle. In condensed matter physics, a Majorana fermion is understood as a quasiparticle whose creation operator coincides with its annihilation operator. The interest in such quasiparticles is due to the fact that they can theoretically be used in qubits for a topological quantum computer, while due to their non-local nature they are less sensitive to the influence of the environment. In one-dimensional systems, one speaks not of Majorana fermions, but of Majorana bound states, which do not move freely in the system, due to which they retain their properties. The possible experimental detection of such objects in combined semiconductor-superconductor nanosystems in a magnetic field requires independent confirmation due to the complexity of detection and the existence of possible alternative explanations.

The Hamiltonian used by Kitaev is very simple, since it contains exchange only between nearest neighbors. This simplicity has enabled to make a number of theoretical predictions of its implementation in solid state physics. Materials in which spin and orbital degrees of freedom are strongly intertwined in the presence of spin-orbit couplings, such as $(Na,Li)_2IrO_3$ and α- $RuCl_3$ are the most likely candidates for implementing Kitaev's theory. In the article [14], within the framework of the extended Kitaev model, the existence of a chiral spin liquid as the ground state „triangle-hexagon" of a spin lattice obtained by replacing each node of the hexagonal lattice by a triangle was rigorously established. The ground state of spin liquid obtained by the authors is one example of a wide class of chiral spin liquids considered earlier by Wen [1]. The symmetry of time reversal is spontaneously broken in it, but other types of symmetry are preserved. Majorana fermions and local gap magnetic fluxes (vortices) were considered as excitations of the ground state of the chiral QSL. It was found that the creation of vortices pairs requires energy, so the vortices do not move along the lattice, even when they interact with each other. Each vortex carries a Majorana mode with zero excitation energy. According to [14], a pair of zero-energy Majorana modes on two well-separated vortices creates one complex mode with zero excitation energy. In the article [15] to describe the ground state of QSL on a hexagonal lattice, the extended Heisenberg-Kitaev $H_{\mathrm{HK}} + h + \kappa$ with an additional perturbation in the form of a three-spin exchange κ in a magnetic field h. It is shown that the three-spin ring exchange κ destroys the time reversal symmetry and promotes the transition of the zero-gap QSL into a gap topologically ordered phase in very weak magnetic fields not only in the Kitaev limit ($\alpha = 1$), but also for all values of the coupling parameter α in a wide range of values $0.8 \le \alpha \le 1$. It is assumed that even in a small external magnetic field ($h \to 0$), QSL with ring exchange should be in a gap state with topological order. According to the constructed ($\kappa - h$) magnetic phase diagram, there is a single phase transition that separates the topologically ordered state of QSL from the fully polarized state in high magnetic fields h. In the Kitaev limit, this transition occurs at the critical field strength of $h_c \approx 0.072$ and remains almost constant as α decreases. It is interesting to note that the critical field h_c initially grows continuously with increasing κ, but then saturates at some finite value of the three-spin exchange of $\kappa \approx 6$. This saturation of the critical field corresponds to the gap behavior for Majorana fermions in the exact solution at $h = 0$. According to [15], the Majorana fermion energy dispersion $E(k)$ in the topological phase depends essentially on the magnitude of the three-spin exchange. For small $\kappa \gg 1$ the fermion has a gap $E_g \approx \sqrt{3}\kappa$, but for large $\kappa \gg 1$ the fermion gap $E_g \approx 2$ is constant and does not depend on the value κ. The field dependences of magnetization $M(h)$ and the derivatives dM/dh are numerically calculated in the range of magnetic fields $0 \le h \le 0.48$ for the values of the exchange interaction $\kappa = 0,\ 0.9,\ 6,\ 30$. According to the results obtained, the field dependences of magnetization $M(h)$ near the transition to a state with a topological order have the form of thresholds, the shape and magnitude of which essentially depend on κ. In the Kitaev limit ($\alpha = 1$), this transition occurs at a critical value of the external magnetic field strength of $h_c \approx 0.072$, which remains constant as the coupling parameter α decreases. It is interesting to note that the phase transition induced by an external magnetic field from a topologically ordered state to a polarized state can be continuous, as well as a weakly expressed first-order phase transition. Since the strength of the critical magnetic field required for destruction of the topological phase is determined by the MF gap at $h = 0$, the value of h_c first increases with the growth of the exchange, and then reaches saturation at large values κ. The dependences of the number of vortices (magnetic fluxes) in the ground state of QSL on the intensity of the external magnetic field, which corresponds to the number of areas with an unusual flux were also constructed. Below the critical magnetic field ($h < h_c$), there are no vortices in QSL due to the presence of a gap for vortices in the state in which there is no confinement. However, during the phase transition to the polarized state, vortices appear due to the compression of QSL, and their number in the ground state rapidly increases with the field.

Thus, the nature of the phase transition can be formulated in terms of the confinement-deconfinement transition of the non-Abelian gage field, which is related to the confinement-deconfinement phase transition in the Abelian discrete gage theory [16–19]. According to the theory, the flux condensation leads to the confinement of the gage field [20]. In this article, we study the nature of two types of features in the field dependences of magnetization $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$, which appear near zero magnetic field H during sample remagnetization in ZFC- and FC-measurement modes in the field range ± 5 kOe at 4.2 K. Appearance of these features of sample magnetization near $H = 0$ is explained by the excitation of a 2D-continuum of fermion pairs and 1D-fragments of charge/spin density waves in the process of sample remagnetization within the framework of existing models of the chiral QSL phase transition into a gap state with a topological quantum order.

2. Experimental procedure

Samples of self-doped $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ manganites were grown from high-purity lanthanum, samarium, and electrolytic manganese oxides taken in a stoichiometric ratio. Synthesized powder was pressed under 10 kbar into disks 6 mm in diameter and 1.2 mm in thickness and sintered in air at a temperature of 1170°C for 20 h with subsequent temperature reduction at a rate of 70° C/h.

According to the results of X-ray studies, the obtained pellets were single-phase ceramics. X-ray diffraction studies were carried out at 300 K using a DRON-1.5 diffractometer in Ni$K_{\alpha 1+\alpha 2}$ radiation. The lattice symmetry and parameters were determined by the position and the nature of splitting of reflections of the perovskite-type pseudo-cubic lattice. The field dependences of magnetization were obtained in the ZFC- and FC-measurement modes dc of magnetization in the range of fields $-5\,\mathrm{kOe} \leq H \leq 5\,\mathrm{kOe}$ at 4.2 K using a magnetometer.

3. Experimental results and discussion

3.1. Features in the dynamic spin structure factor of a chiral QSL under excitation of a 2D-continuum of quasiparticles in the form of Majorana fermions and local fluxes Z_2 of a gage field

It can be seen from Fig. 1 that in the remagnetization processes of $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in ZFC- and FC-measurement modes, sample magnetization $M(H)$ grows linearly with increasing magnetic field strength in a wide range of magnetic fields, which is typical for paramagnets and antiferromagnets, but has clearly pronounced unusual features near zero field, caused by the appearance of additional „supermagnetization" upon QSL transition into a phase with a topological order. The strong field hysteresis is also unusual, the width of which in the FC-measurement mode is much larger than in the ZFC-mode.

As can be seen in Fig. 2, in the ZFC-measurement mode during remagnetization near the critical fields $H_{c1} \approx -50$ Oe and $H_{c2} \approx 50$ Oe thresholds close in shape are formed, respectively, for the magnetization isotherm — *1* and demagnetization isothem — *2*, followed by a smooth drop of sample „supermagnetization" to zero. Fig. 2 clearly

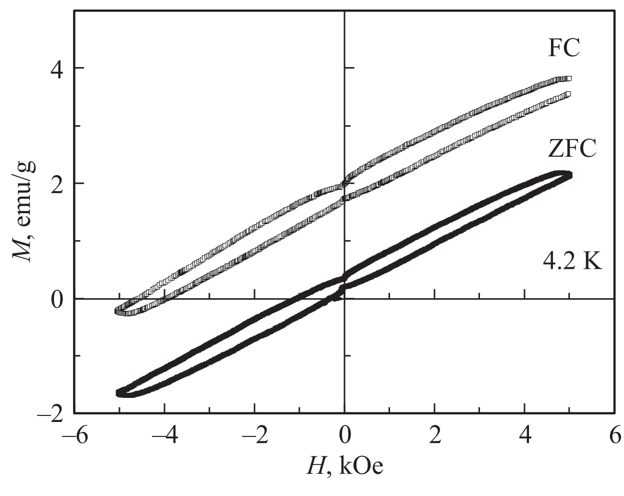

Figure 1. Field dependences of magnetization $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in ZFC- and FC -measurement modes during sample remagnetization in the range of magnetic fields $-5\,\mathrm{kOe} \leq H \leq 5\,\mathrm{kOe}$.

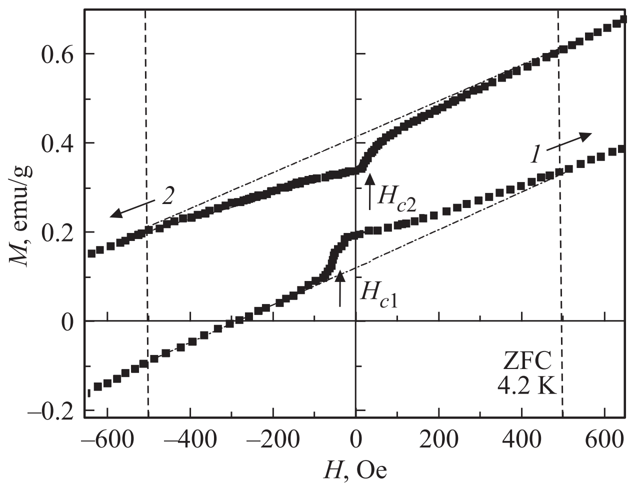

Figure 2. Formation of a hillock-like asymmetric feature of $M(H)$ plots in ZFC and measurement mode in the range of magnetic fields ± 500 Oe with increasing magnetic field strength in the positive direction (isotherm *1*) and in the negative direction (isotherm *2*).

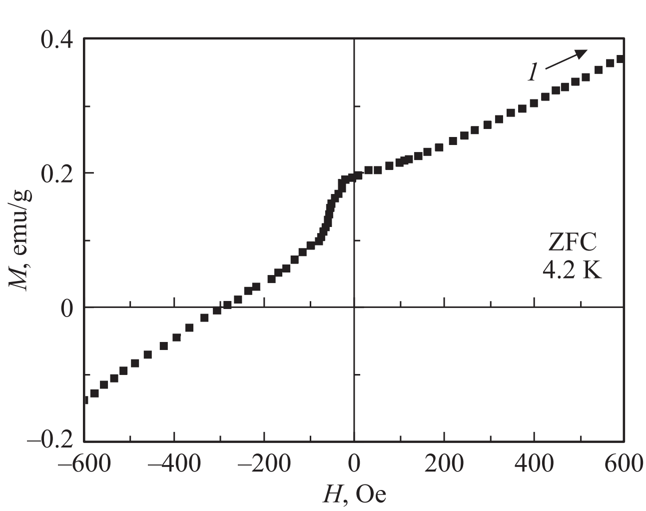

Figure 3. Magnetization isotherm *1* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in ZFC-measurement mode in the range of magnetic fields ± 600 Oe.

shows how the hillock-like asymmetric feature of the $M(H)$ plots is formed in the range of magnetic fields ± 500 Oe as the magnetic field strength increases in the positive direction. It is clearly seen that this unusual feature is a superposition of a relatively narrow threshold feature of magnetization near the critical field $H_{c1} \approx -50$ Oe of the QSL phase transition to a state with a topological order, which is superimposed on a smooth drop of „supermagnetization" to zero near magnetic fields of ± 500 Oe in the form of a tail. Figure 3 shows that the dominant contribution to the growth of „supermagnetization" in the *1* isotherm with increasing field is made by a clearly pronounced positive jump in the magnetization near the field $H_{c1} \neq 0$. However, according to Fig. 4, there is also an additional contribution

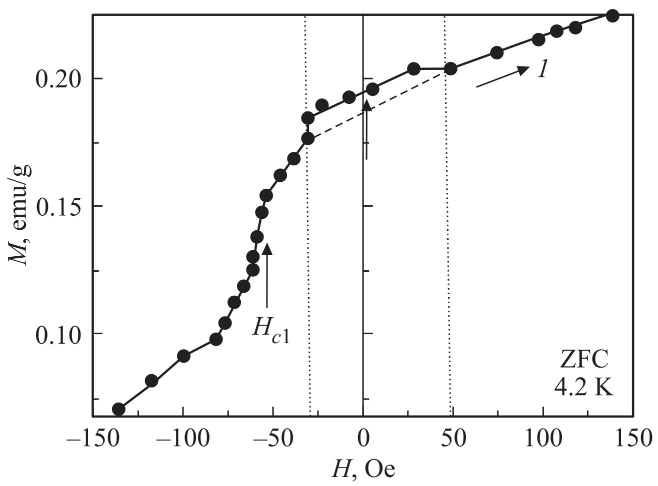

Figure 4. Fragment of the magnetization isotherm *1* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in ZFC-measurement mode in the range of magnetic fields ±150 Oe.

to magnetization of the sample in the form of a plateau located in a narrow range of fields near the zero magnetic field.

Thus, the asymmetric hillock-like feature of the $M(H)$ plots obtained by us in the range of magnetic fields ±500 Oe with increasing magnetic field strength in the positive direction (isotherm *1*) consists of two contributions that differ in shape and intensity: an asymmetric hillock-like feature in the field range ±500 Oe and a narrower plateau-like feature near $H = 0$. A similar asymmetric hillock-like feature of the $M(H)$ plots is also formed with a decrease in the field during the reverse cycle of remagnetization, shown on the *2* isotherm in Figs 2, 5, 6. It is clearly seen in Figs 5, 6 that the magnetic response of the spin system during sample demagnetization near zero field has the form of a threshold drop of „supermagnetization" with increasing field near the critical value of $H_{c2} \approx 50$ Oe. Critical fields, form and intensity of additional contributions of low-energy excitation of quasiparticles to the total sample magnetization on two isotherms differ significantly, that is, there is a strong hysteresis in the sample magnetization — demagnetization processes. Note also the unusual asymmetry of the $M(H)$ curves for the left ($H < 0$) and right ($H > 0$) halves of the field dependences of the QSL magnetization in the *1* and *2* isotherms, which indicates the violation of the mirror symmetry of the sample magnetization upon reversal of the sign of the external magnetic field.

Thus, according to the field dependences of magnetization $M(H)$ obtained in this article, measured in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in ZFC-measurement mode at a temperature of 4.2 K, with an increase in external magnetic field strength, a second-order threshold quantum phase transition of the QSL to a state with a topological order occurs, followed by a smooth transition to the polarized phase. As a result of this transition, in the remagnetization isotherms of *1* and *2* samples, near the value $H = 0$, hillock-like features of magnetization $M(H)$ are formed, the shape and intensity of which significantly depend on the direction of the magnetic field growth. These features are typical for the low-energy excitation of a wide 2D-continuum of Majorana fermions during the phase transition of chiral QSL to a state with a topological order, which was considered earlier in articles [13–28]. An important consequence of Kitaev's model for QSL [13] is the splitting of electrons into a set of unusual quasiparticles —Majorana fermions, which leads to the appearance of chiral spin liquids of several Majorana fermion bands in the spectrum of ground state magnetic low-energy excitations, the width of which is significantly depends on the magnetic gage fluxes and external perturbations. More recently, evidence of this splitting has been found in α-$RuCl_3$ using neutron scattering techniques, which were explained in [21]. In the artile [21] a theoretical

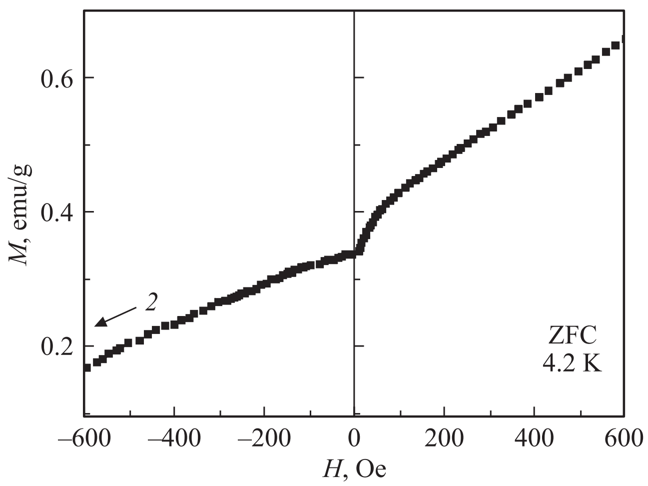

Figure 5. Demagnetization isotherm *2* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in ZFC-measurement mode in the range of magnetic fields ±600 Oe.

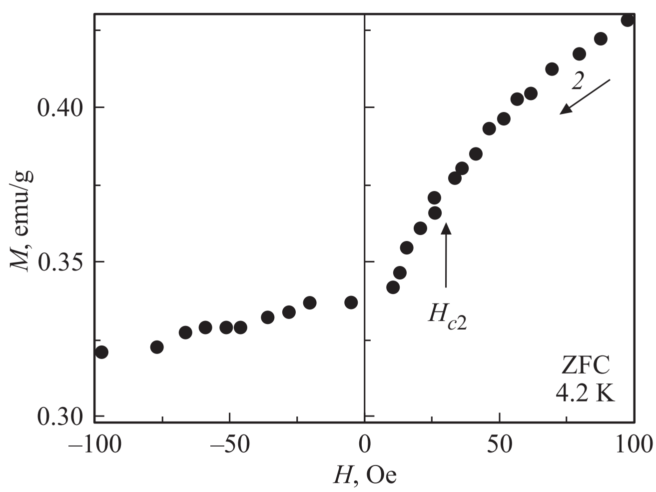

Figure 6. Fragment of the demagnetization isotherm *2* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in ZFC-measurement mode in the range of magnetic fields ±100 Oe.

study of the dynamic structure factor $S^{ab}(\mathbf{q}, \omega)$, a two-dimensional quantum spin liquid in the zero gap and gap phases, was carried out within the framework of Kitaev's model for the hexagonal lattice of spins. The existence of unusual hillock-like features of the structure factor caused by the excitation of the continuum of Majorana fermions and the appearance of gage fluxes is shown. It was found that both in the gap and zero gap phases of QSL, the response vanishes at values of the excitation energy ω below the gap value of Δ, which is directly related to the appearance of the gage field. A similar gap was also found in the modified Kitaev model in the spectrum of circular excitation [22].

For excitation energies higher than the gap energy Δ, non-zero fermion contributions to the structure factor $S^{ab}(\omega)$ arise only from excited states with a parity opposite to that of the ground state. As a result of this, two modes of excitation of Majorana fermions are implemented — mode I and mode II. Mode II contains a spiky response, while mode I has no spiky response. In case I, the ground state of QSL has an odd number of excitations, while in case II, magnetic excitations are either absent or their number is even. In mode I, a broad hillock-like magnetic response to external excitation is observed, while, in case II, the response should be narrow. The responses may overlap or be separate. For case I, single-particle excitations dominate in the energy-wide response; therefore, it manifests itself only within the band width of material fermions.

In the article [23] exact results are obtained for the structure factor in the gap and zero gap, Abelian and non-Abelian phases of QSL. The structure factor has features in the appearance of quasiparticles in the form of Majorana fermions and fluxes Z_2 of the gage field. In addition to a broad hillock-like continuum of fractional spin excitations, the authors found spiky features in the magnetic response. These spiky features arise from excited states containing either only static magnetic fluxes and no mobile fermions, or from excited states in which fermions are closely coupled to fluxes. The structure factor is significantly different in the Abelian and non-Abelian QSLs. Bound fermion fluxes-composites appear only in the non-Abelian phase. The main feature of the dynamical structure factor at the isotropic point of the non-Abelian phase $(Jx = Jy = Jz)$ is the presence of a pointed δ-component caused by Majorana fermions coupled to flux pairs and a broad hillock-like component caused by fermion continuum excitation. The beginning of the broad feature of the magnetic response corresponds to the edge of the Majorana fermion band. It should be noted that the width of the response of free fermions practically coincides with the width of the calculated function of the density of states $N(\omega)$ of Majorana fermions in the phase with zero flux.

According to Kitaev's model, a spin system with $S = 1/2$ is occupied by nodes in a hexagonal lattice. The spins interact via an anisotropic Ising exchange J_a between nearest neighbors, where three directions denoted as $a = x, y, z$ allocate three valence bonds for a given lattice position. Kitaev's model is supplemented in [23] by a three-spin exchange interaction created by a weak external magnetic field. Circular three-spin exchange interactions break the time symmetry and create a gap in the spectrum of Majorana fermions, which leads to an increase in non-Abelian excitations [24]. The first term of the Hamiltonian of the extended Kitaev model $\hat{\tilde{\mathrm{H}}} = -\sum_{nn} J_a \hat{\sigma}_i^a \hat{\sigma}_j^a - K \sum_{nnn} J_a \hat{\sigma}_i^a \hat{\sigma}_j^c \hat{\sigma}_k^b$ used in the article [23], sums (nn) Ising exchange interactions in a spin system between two neighboring i, j lattice nodes of spins in terms of Pauli matrices $\hat{\sigma}_j^a$. The additional term of the Hamiltonian, taken with a coefficient K, describes (nnn) exchange interactions between three spins $\hat{\sigma}_i^a \hat{\sigma}_j^c \hat{\sigma}_k^b$ combined with each pair of links $\langle ij \rangle_a$ and $\langle jk \rangle_b$ for a node j of the crystal lattice. For $K = 0$, there can exist two stable phases known in the literature as zero gap and gap Abelian quantum spin liquids. At $K \neq 0$, all these phases acquire a gap, and the former zero gap excitations for the QSL state become non-Abelian. In all phases, the independent degrees of freedom are Z_2 gage fluxes located in the crystal lattice cells and dynamic Majorana fermions located at the lattice nodes. The time evolution of Majorana fermions is generated by a Hamiltonian whose form is determined by a special configuration Z_2 of the gage field. As noted in [23], different representatives of the family of Kitaev Hamiltonians correspond to a set of qualitatively different responses. All these responses have in common that the found spin correlations are ultrashort [25]: the structure factor contains the contributions of the correlators of only local or nearest neighbors that have the same spin components. This is a consequence of the static nature of the resulting Z_2 gage fluxes. Therefore, only the dependences of the dynamic structure factor $S(\omega)$ on the excitation frequency $(q = 0)$ were presented in the article. In all cases, the dynamic response has a gap, regardless of the existence of a gap in the excitation spectrum of the quantum spin liquid. The minimum gap Δ corresponds to the case of the flux pair formation in QsL in adjacent cells of the crystal lattice. The plot of one of the broad components of the magnetic response $S(\omega)$ corresponds to the excitation of Majorana fermions $(q = 0)$. Its low-energy start corresponds to a Δ gap in the phase with zero gap Majorana excitations, while the end is in the gap Majorana phase. As shown in the article, this broad response arises when one or two fermions are excited, depending on the spin component and the parameters of the Hamiltonian. In the case $J_x/J_z = J_y/J_z$ there are only two response components — $S^{zz}(\omega)$ and $S^{xx}(\omega) = S^{yy}(\omega)$. For the Abelian states of QSL, it was found that in the isotropic model the response $S^{aa}(\omega)$ is non-zero at excitation energy higher than the energy Δ required to create flux pairs. This dominant contribution arises from single excitations of Majorana fermions, but the tail is pulled to higher energies. Although the Majorana fermion band width mainly determines the magnetic response of the spin system to an external perturbation, the dependence of the response intensity is not simply related to the fermion state density function, since the response involves the multiplication of Majorana fermions in the presence of flux pairs. Nevertheless, it reflects such characteristic features of the Majorana state density function as, for example, the Van Hove singularity.

According to [26], the broad hillock-like response of neutron scattering by a spin system in QSL has several different causes. First, this is scattering, in which any movement of spins with a wave vector $q = 0$ requires creation of many magnetic excitations. Secondly, this is not a simple excitation of magnons, since only fractional magnetic excitations participate in it, which occur simultaneously, and thus the scattering process is multiparticle. Thirdly, -when describing the low-energy spectrum, quasiparticles are not in the first place, so the $\omega(q)$ dispersion relation is not essential in principle. In the case of Z_2 spin liquid, in which the triplet excitation decays into a pair of $S = 1/2$ spinons, fractional magnetic excitations are considered as magnetic moments within the framework of the quantum dimer model. In order to be observable, each individual spinon must behave like a coherent free particle in the limit of low excitation energies. However, it is still unclear in what energy range such long-lived well-defined fractional particles will exist. The case of symmetric splitting into two particles is a simplified scenario. -Apparently, many other quasiparticles appear additionally, which enables to approach particle cartridge structures with a wide variety of different fragmentation schemes considered by Wen in [5]. Fractional quasiparticles are not required to have a wide continuous spectrum. Instead, they can form bound or localized states. There is a possibility of quantum chirality deconfinement at the bicritical point of the phase diagram, which separates two phases with the presence of c of quantum spin liquid excitations [27,28]. Deconfinement of bound states of fractional particles is accompanied by the energy consumption required for their decoupling. This does not exclude the possibility of the formation of new discrete composite states with a finite bond energy. One such case is fragmentation with a very large excitation asymmetry, in which the flux can be very heavy and slow moving, i.e. have a very small zone width. Thus, it can interact with an arbitrary number of moments with almost constant energy and thereby produce preservation of extremely inefficient moments. This leads to broad features of particle excitation, which is typical for photons [21,23].

3.2. Formation of bosons in the form of 1D-charge/spin density waves in systems of AFM spin chains induced by confinement of spinon pairs

Of great interest are the remagnetization isotherms obtained by us in the FC-measurement mode, from which it can be seen that when the sample is remagnetized near zero magnetic field, plateau-like features of magnetization are formed, which are characteristic of excitation of 1D-fragments of zero gap charge/spin density waves.

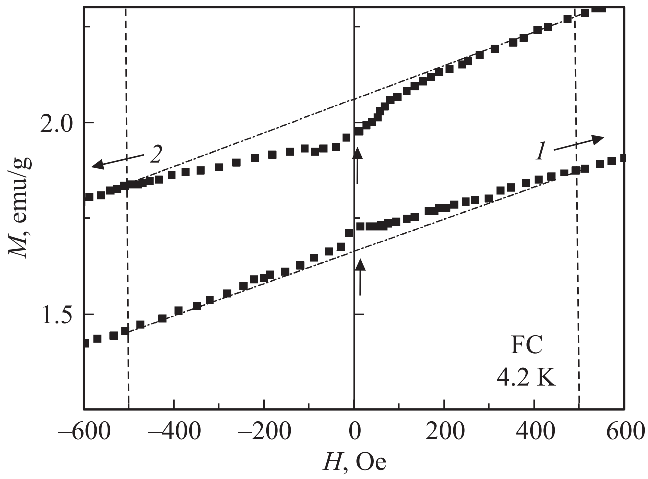

Figure 7. Formation of plateau-like features of $M(H)$ plots in FC and magnetization measurement mode in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in the range of magnetic fields ± 600 Oe with increasing magnetic field strength in the positive direction (isotherm *1*) and in the negative direction (isotherm *2*).

As can be seen in Fig. 7, during remagnetization of the $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ sample at 4.2 K, near the zero magnetic field, similar in shape, relatively narrow plateau-like features of the *1* and *2* isotherms with different widths and intensities are formed. These unusual features of „supermagnetization“, which arise when the **H** vector direction is inverted, are superimposed on a smooth drop in the magnetization to zero with increasing field strength near magnetic fields ± 500 Oe. Figures 8, 9 show that during sample magnetization with increasing H (isotherm *1*), near $H = 0$ a weak plateau-like jump of „supermagnetization“ with a width of 80 Oe is formed in the form of a hillock with a truncated top. Formation of this unusual additional feature of „supermagnetization“ near zero magnetic field manifested itself more clearly in the *2* isotherm during sample demagnetization (Fig. 10−11) in the form of $M(H)$ drop. Appearance of such an unusual feature of the magnetization in the *1, 2* isotherms can be explained by the confinement (deconfinement) of low-energy spinon excitations in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in the form of excitations (destructions) of nanofragments of zero gap 1D-charge/spin density waves, well known in the literature for systems of conducting spin chains in low-dimensional antiferromagnets [29–35]. These collective excitations are superimposed on a wide 2D-continuum of low-energy Majorana fermion excitations during the QSL phase transition to a chiral state with a topological order, which we considered in subsection 3.1.

The mechanism of formation of a gapless mode of fundamental longitudinal oscillations of a system of AFM spin chains in Yb_2Pt_2Pb was studied by high-resolution neutron scattering in strong magnetic fields in the article [29]. Earlier experiments on neutron scattering have demonstrated that $4f$-orbital overlapping imparts unusual magnetic properties to Yb_2Pt_2Pb metal, where orthogonal pairs of Yb ions with pseudospin $S = 1/2$ are located in tetragonal ab planes. It was found that low-energy spinons are the excitations of the magnetic system of spin chains in zero magnetic field. These spinons form a strongly disperse excitation continuum with its width exceeding the energy gap considerably with wavevector $\mathbf{q}_L$ directed along the chains. The boundaries of the two-spinon excitation continuum are set by the extreme values of energy E and linear momentum q of a spinon pair, which are conserved by one particle and one hole with spin $S^z = \pm 1/2$, respectively. In zero magnetic field, chemical potential $\mu = 0$ is in the middle of spinon energy gap Δ_S separating the bands of particles and holes with strong dispersion. An entirely flat continuum of spinon excitations is established for spinons with a perpendicular direction of wavevector $\mathbf{q}_{HH}$. This is indicative of complete uncoupling

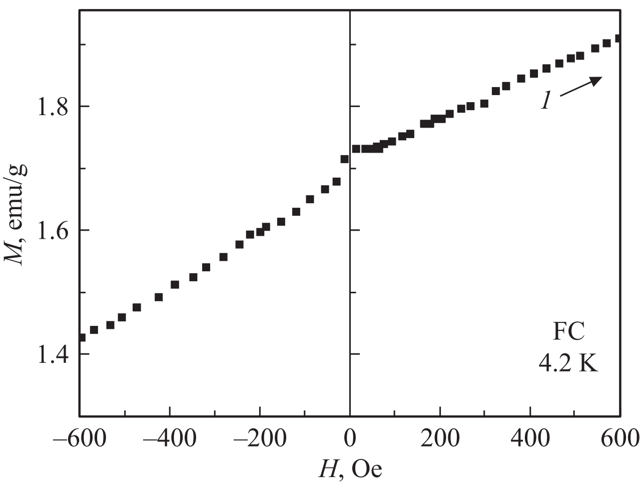

Figure 8. Magnetization isotherm *1* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in FC-measurement mode in the range of magnetic fields ± 600 Oe.

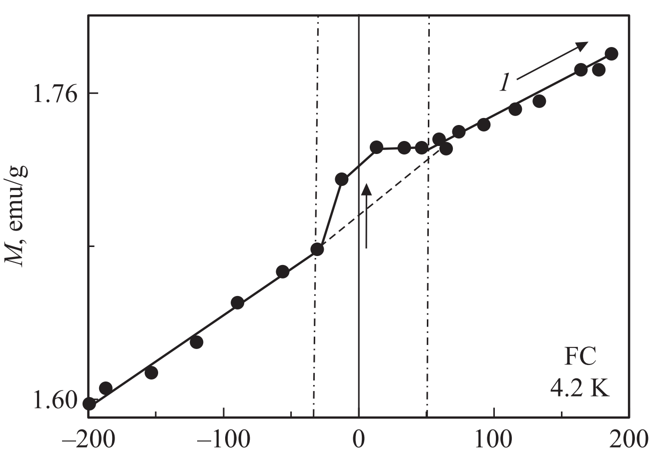

Figure 9. Fragment of the magnetization isotherm *1* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in FC-measurement mode in the range of magnetic fields ± 200 Oe.

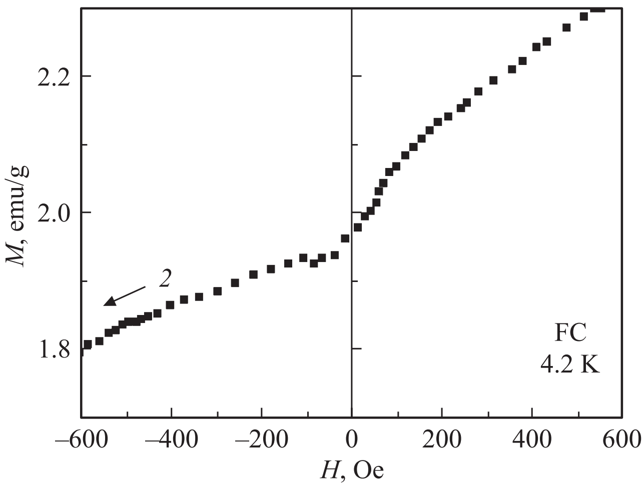

Figure 10. Demagnetization isotherm *2* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in FC-measurement mode in the range of magnetic fields ± 600 Oe.

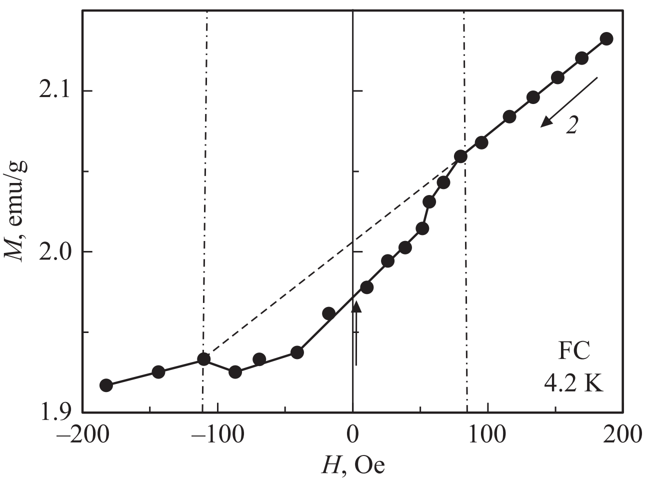

Figure 11. Fragment of the demagnetization isotherm *1* of $M(H)$ in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ at 4.2 K in FC-measurement mode in the range of magnetic fields ± 200 Oe.

of spinons from different chains. The flat dispersion of interchain excitations in zero magnetic field suggests that the interchain interaction at low excitation energies is frozen when spinon gap $\Delta_S \neq 0$. It was found for spinons with wavevector $\mathbf{q}_L$ that an increase in magnetic field intensity within $0 \leq H \leq 30$ kOe is accompanied by a significant alteration of the shape of the spinon excitation spectrum in Yb_2Pt_2Pb. This is attributable to the fact that an increasing magnetic field alters the occupancy of disperse spinon bands with $S^z = \pm 1/2$ due to the shift of chemical potential μ in energy gap Δ_S. The Zeeman interaction with a magnetic field reduces the system potential to $\mu = -g\mu_B H S^z$, which may result in gap closure in sufficiently strong fields. It was established that the potential needed for gap closure and the emergence of holes at temperatures $k_B T < \Delta_S$ is $|\mu| = \Delta_S = 0.095$ meV. This value is attained when the field increases to critical $H_{c1} = 5$ kOe. In the limit of relatively weak fields, a jump in magnetization was observed instead of a linear increase in a magnetic field increasing from zero to H_{c1}. It was demonstrated that gap closure at $H \geq H_{c1}$ results in confinement of spinon pairs, and a gapless mode of fundamental longitudinal oscillations of the system of spin chains arises from this confinement.

The confinement of low-energy excitations (spinons) with fractional quantum numbers in a system of AFM chains of spins $S = 1/2$ coupled by weak exchange interactions in Heisenberg $CaCu_2O_3$ AFM with critical temperature $T_N = 25$ K of AFM ordering was examined in [30]. It is typical of states with a spatially restricted spinon motion for spinons in a system of chains to be coupled by an interaction that becomes stronger with distance. One consequence of this is that such excitations cannot be observed individually. The authors draw an analogy between spinon confinement in a system of AFM chains of spins $S = 1/2$ and similar phenomena known from particle physics, where heavy particles (baryons and mesons) are created due to quark confinement. A simplified pattern of spinon confinement is observed in chains with a significant Ising-like anisotropy of the exchange interaction, where the ground states have a finite Néel order. Spinons then act as domain walls separating two degenerate states with opposite magnetizations. In case of chains with Heisenberg exchange between spins, the situation is different, although domain walls are always created in pairs in an individual chain, no energy is lost when they are moving relative to each other. Therefore, in this case spinons in an individual chain are not coupled into pairs of spins $S = 1/2$ by strong attraction. In contrast to the Ising limit, only two types of chain excitation (triplet and singlet) exist at the point with zero anisotropy of exchange interaction between spins. Excitations in an individual chain (spinons) with spin $S = 1/2$ are confined even by an infinitesimal interchain coupling. The majority of spin ladders studied earlier do not exhibit this effect, since a strong interchain interaction suppresses the excitation of spinons at all excitation energies. The article presents the results of studying neutron scattering for a system of weakly coupled chains in spin ladders in $CaCu_2O_3$ cuprate, which has orthorhombic symmetry *Pmmn*. The crystal structure consists of CuO-layers perpendicular to the c direction; the spin chains lie in the ab-planes along the b-direction and are shifted by half the lattice cell along the a-axis. Ladders are coupled by several weak interactions. Inside the ab-planes, the Cu^{2+} ions on adjacent ladders are coupled to each other via Cu−O−Cu-bonds. Just as the other planar cuprates, $CaCu_2O_3$ supports four-spin exchange interaction J_{cyclic} that couples four copper ions, which form rectangular plaquettes in ab-planes. According to the results of calculations performed by other research groups, the primary exchange constants are $J_{\text{leg}} = -147$ meV; $J_{\text{rung}} = -15$ meV; $J_{\text{cyclic}} = 4$ meV, where J_{leg}, J_{rung} are the leg and rung exchange constants, respectively. The region between two spinons (domain walls) in a chain consists of inverted spins. If this chain is coupled antiferromagnetically with another chain in a spin ladder, these inverted spins lose energy (compared to the case when they are parallel to spins of the neighboring chain). This energy loss, which is proportional to the distance between spinons, incudes their confinement. In the article it was found that the magnetic properties of the system of chains in ladders at high excitation energies are similar to those of uncoupled individual chains. The neutron-scattering signal corresponds in this case to scattering typical of an individual chain of spins $S = 1/2$ with an interchain leg exchange constant $J_{\text{leg}} = -162$ meV. At the same time, integral spin excitations typical of strongly coupled chains are dominant at low energies of excitation of chains by neutrons. Thus, weakly coupled ladders in $CaCu_2O_3$ differ significantly from the well-studied strongly coupled spin ladders that always remain in the strong confinement regime and support only magnon excitations ($S = 1$).

In the artile [31] system of Ising-like XXZ weakly coupled antiferromagnetic chains in $SrCo_2V_2O_8$ in the external magnetic field was studied using high-resolution terahertz spectroscopy. A series of excitations with characteristic Zeeman splitting in the external magnetic field was observed at low temperatures. The authors identified these magnetic excitations as confined spinon-pair excitations. The absorption spectrum measured in light transmission through the sample at a temperature of 6.5 K (slightly higher than $T_N = 5$ K) within an energy interval of 1.3−6 meV contains intense absorption line E_1 of light with an oscillation frequency around $\omega = 0.35$ THz and a subsequent series of absorption lines which were abruptly reduced to zero intensity when the frequency increased to $\omega = 1.2$ THz. These absorption lines were not observed when the ac magnetic field of light wave $h(\omega)$ was directed parallel to the spins in chains along axis c of the crystal lattice. This is indicative of the magnetic nature of lines of the discovered discrete light absorption spectrum, which is governed by the excitation of spinon pairs. The measured dependence of intensity of these lines on the external dc magnetic field verified this conclusion. In a single Ising chain, two spinons are created by a spin flip. Spinons with spin-1/2, may move freely along the chain by subsequent spin flips without any loss of energy. This results in the formation of a highly degenerate first excited state with energy J, which is equal to the intrachain nearest-neighbor exchange interaction. A finite transverse component of the intrachain exchange interaction lifts the degeneracy, leading to an excitation continuum of two spinons that propagate independently along the chain. In the presence of weak interchain interactions, two spinons are coupled, since their separation frustrates the interchain exchange interaction. The interchain interaction acts as an attractive potential $V(z)$ for spinons that is proportional to distance z between them. Thus, the weak interchain interaction in the reason behind linear confinement of spinons into coupled pairs. Potential $V(z)$ induces quantization of the excitation continuum of the system of chains in $SrCo_2V_2O_8$ into discrete levels E_i of coupled spinon pairs.

The dynamics of excitation of spinon pairs in an Ising-like antiferromagnet $BaCo_2V_2O_8$ at temperatures below $T_N \approx 5.5\,K$ was studied by inelastic neutron scattering in [32]. The authors note that a one-dimensional antiferromagnet is of special interest, since quantum fluctuations in a one-dimensional system of spins melt the classical long-range Néel order. The ground state becomes disordered in this case. The excitation spectrum of this disordered ground state of a 1D-system is a continuum of many excitation pairs with spin $S = 1/2$ (spinons) similar to domain walls in an Ising magnet. However, a one-dimensional system of spins may become AFM-ordered at very low temperatures in the presence of even a very weak coupling between chains. Three possible spin states $S = \pm 1$, 0 for spinon pairs then transform into two transverse oscillation modes and a third (collective) excitation that corresponds to fluctuations parallel to the ordered momentum (i.e., longitudinal oscillations). An unusual spectrum of spin excitations, which was explained in terms of spinon confinement induced by the interchain interactions, was discovered in the article at measurement temperature $T = 1.6\,K$ below T_N. These excitations consist of two types of alternating series with transverse and longitudinal polarizations. Similar series of excitations of Ising spin chains have been observed earlier in $CsCoCl_3$, $CsCoBr_3$ and $CoNb_2O_6$. Longitudinal oscillations in chains correspond to longitudinal fluctuations of the ordered momentum. The obtained experimental data revealed that $BaCo_2V_2O_8$ with moderate Ising anisotropy and significant interchain interactions satisfies well the requirements for observation of long-lived longitudinal oscillations of a 1D-system of spins. Summing up the results of analysis of experimental data, the authors conclude that excitations in $BaCo_2V_2O_8$ at temperatures below T_N are quantized due to the weak interchain interaction and consist of two alternating series of transverse and a remarkably strong longitudinal Zeeman ladders. It is assumed that longitudinal modes are stabilized owing to moderate Ising anisotropy, which prevents their decomposition into discrete gap transverse modes.

Magnetic excitations in $BaCo_2V_2O_8$ were studied in [33] by elastic and inelastic neutron scattering at temperature $T = 3.5\,K$ as functions of intensity H of a transverse dc magnetic field within the $0-10\,T$ interval. The field was oriented perpendicular to the system of AFM-chains with Ising-like exchange along axis c of the crystal lattice. Earlier studies into the influence of a longitudinal magnetic field on the magnetic properties of $S = 1/2$ 1D-antiferromagnet with Ising-like anisotropy have revealed that a quantum phase transition from an ordered state with long-range AFM ordering to a disordered phase similar to a spin liquid is likely to occur in the process of H growth. This transition is attributable to the development of a longitudinal spin correlation incommensurate with the crystal lattice in an increasing field. A finite magnitude of interchain exchange interactions induces the emergence and growth of a phase with a long-range longitudinal spin density wave and initiates variations of its incommensurability with the lattice. The influence of transverse magnetic fields on phase transitions in similar systems of spin chains has also been examined. It was demonstrated that a transverse external magnetic field also affects the magnetic order in $BaCo_2V_2O_8$ (e.g., suppresses it at values $H \sim 1\,T$). We are mostly interested in the results of examination of the influence of transverse fields on the spinon spectrum in [33]. It was found that the peak features of series of spinon excitations split gradually as the external magnetic field increases. It was demonstrated clearly that the behavior of the magnetic excitation spectrum in $BaCo_2V_2O_8$ in a magnetic field is characterized well by a model based on the theory of an antiferromagnetic XXZ chain of spins with $S = 1/2$. This model predicts that the dynamic magnetic structure factor of the spin component along the chain increases with H, and well-marked incommensurate spin correlations emerge. A radical modification of quantum excitations in $BaCo_2V_2O_8$ in a transverse field at H exceeding the critical field value was discovered in [34] in neutron-scattering experiments. It was demonstrated that this modification is induced by the quantum phase transition between two different types of soliton-like topological excitations. The process of spinon confinement in a quasi-one-dimensional anisotropic Heisenberg antiferromagnet ($SrCo_2V_2O_8$) was studied later in [35] by inelastic neutron scattering within a wide temperature interval above and below critical temperature $T_N = 5.2\,K$ of the phase transition to the AFM-state. The spinon continuum observed at $T = 6\,K$ in the form of an intense peak feature smeared within a wide interval of spinon excitation energies is in good agreement with the theoretical prediction for the transverse dynamical structure factor of a Heisenberg XXZ-chain at zero temperature. At temperatures below T_N, pairs of spinons are confined, and two series of meson-like bound states with longitudinal and transverse polarizations, which correspond to the propagation of excitation of an XXZ chain by neutrons in longitudinal and transverse directions, are observed. It was found that the broad peak feature transforms into two series of narrow sharp peaks when temperature drops to $T = 1.5\,K$. The width and the intensity of these narrow peaks depend strongly on the excitation conditions. The results of our study of $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in the FC-measurement mode at a temperature of 4.2 K are in good agreement with the general picture of the formation of zero gap bosons in the form of fragments of 1D-charge/spin density waves, within the framework of existing confinement models of spinon pairs.

4. Conclusion

According to the extended Heisenberg–Kitaev models in quasi-two-dimensional AFMs with three-spin exchange interaction, Majorana fermions can appear in the spectrum of low-energy QSL excitations as a quantized gas of fractional excitation pairs with $S = 1/2$ even in very weak external magnetic fields. It is possible to change the density of Majorana fermions controlled by an external magnetic field without creating electrons in the conduction band or holes in the valence band. For this, only a slow change in the direction (turn) of the magnetic flux of the gage field caused by an external magnetic field is sufficient.

According to the results obtained in this article, the remagnetization process of a quantum spin liquid in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ causes a spin flip controlled by an external magnetic field, which leads to the appearance of magnetic flux pairs in neighboring areas and dynamic rearrangement of Majorana fermions in a magnetic gage field. Results of measuring the field dependences of the magnetization $M(H)$ of quantum spin liquid in frustrated manganites in the $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in ZFC-measurement mode at 4.2 K are in good agreement with the well-known asymmetric hillock-like shape of the QSL magnetic response in frustrated quasi-two-dimensional antiferromagnetic materials near a second-order phase transition to a chiral state with a topological order induced by an increase in an external magnetic field. The transition is accompanied by a threshold increase in the concentration of excitations in the form of Majorana fermions and local magnetic fluxes. The transition occurs in weak critical magnetic fields H_c close to zero and is accompanied by subsequent continuous polarization of the spin system with an increase in the strength of the external magnetic field in the range ± 500 Oe. However, critical fields, form and intensity of additional contributions of low-energy excitation of quasiparticles to the total sample magnetization on two isotherms $M(H)$ differ significantly, that is, there is a strong field hysteresis in the sample magnetization-demagnetization processes. Note also the unusual asymmetry of the „supermagnetiation" $M(H)$ curves for the left $(H < 0)$ and right $(H > 0)$ halves of the field dependences of the QSL magnetization in the *1* and *2* isotherms, which indicates the violation of the mirror symmetry of the sample magnetization upon reversal of the sign of the external magnetic field to the opposite one. Thus, when the sample is remagnetized at 4.2 K in the ZFC-measurement mode near the zero field, wide asymmetric hillock-like features of remagnetization isotherms with low (zero) excitation energy are formed, the width and shape of which significantly depend on the direction of magneticc field growth, which is typical for the low-energy excitation of the 2D-continuum of Majorana fermions and local fluxes Z_2 of the gage field during the phase transition of a chiral QSL to a state with a topological order. At the same time, when the sample is remagnetized in the FC-mode near zero magnetic field, narrow plateau-like features of the magnetization are formed, which are characteristic of the excitation (destruction) of 1D-fragments of zero gap charge/spin density waves. These unusual features of „supermagnetization", which arise when the **H** vector direction is inverted, are superimposed on a smooth drop in the magnetization to zero with increasing field strength near magnetic fields ± 500 Oe. Appearance of such magnetization features can be explained by confinement (deconfinement) of spinon pairs in $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in the form of excitation (destruction) of nanofragments of zero gap 1D-charge/spin density waves well known in the literature for systems of conducting spin chains in low-dimensional antiferromagnets. These zero gap 1D-collective excitations coexist at 4.2 K with a wide 2D-continuum of low-energy Majorana fermion excitations during the QSL phase transition to a chiral state with a topological order in the field interval ± 500 Oe. Thus, the results of our study of $La_{0.15}Sm_{0.85}MnO_{3+\delta}$ in the FC-measurement mode at a temperature of 4.2 K are in good agreement with the picture of the formation of zero gap bosons in the form of fragments of 1D-charge/spin density waves, within the framework of existing confinement models spinon pairs in a system of conducting AFM spin chains coupled by a weak exchange interaction.

Conflict of interest

The authors declare that they have no conflict of interest.

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